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Electronic properties of CePd$_2$Si$_2$ under pressure

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Resistivity measurements were performed up to pressures of 10 GPa and down to temperatures of 30 mK to study the pressure-induced superconductivity of the antiferromagnetic compound CePd$_2$Si$_2$. A large superconducting domain is found in the range 2–7 GPa. Non-Fermi-liquid properties are observed for the pressure corresponding to optimal superconductivity at around 5 GPa where the transition temperature T_c^{onset} equals 520 mK. The high value of the initial slope of the superconducting upper critical field (about -10 T/K) is indicative of heavy fermion superconductivity.

I. INTRODUCTION

Heavy fermion (HF) compounds provide the opportunity to study the anomalous properties occurring at the quantum critical point (QCP) of three-dimensional itinerant antiferromagnets.[1] At the QCP, the Néel temperature T_N vanishes as a function of a control parameter, experimentally achieved by alloying or applying external pressure. Breakdown of the Fermi-liquid theory is expected at the QCP. The corresponding so-called non-Fermi-liquid (NFL) behavior observed may also be characteristic of large crossover regions delimited by the competition between thermal and quantum fluctuations.[2,3] Most surprising in this context is the emergence of superconductivity near the QCP pointing towards the probable magnetically mediated pairing mechanism.

Up to very recently and despite a wealth of experimental effort, CeCu$_2$Si$_2$ was the only known ambient pressure Ce-based HF superconductor.[4] This compound is a member of the extensively studied CeM_2T_2 family (space group I4/mmm) where M is a transition metal and $T=$Si, Ge. A magnetic state is stabilized above a critical volume of the unit cell obtained for different combinations of M and T elements. This gives rise to various kinds of antiferromagnetic order within this family of compounds.[5] Having in mind that the application of pressure counterbalances such volume effects by driving these compounds towards a nonmagnetic state, CeCu$_2$Ge$_2$ (Ref. 6) and CeRh$_2$Si$_2$ (Ref. 7) were subsequently found superconductor under pressure at the QCP.

Recently, focus was made on the isoelectronic compounds CeNi$_2$Ge$_2$ and CePd$_2$Si$_2$. For the former paramagnetic compound, traces of superconductivity were found by several groups both at ambient pressure[8] and above 1.5 GPa.[9–11] For the latter antiferromagnetic compound CePd$_2$Si$_2$ [with $T_N=10$ K and a staggered magnetic moment $m=0.62\mu_B$ (Ref. 5)], only the Cambridge group found so far pressure-induced superconductivity at around 2.7 GPa,[12,13] as opposed to other experimental works.[14,15] In this paper, we confirm the pressure-induced superconductivity of CePd$_2$Si$_2$ and give details on the electronic properties associated with the superconducting phase.

II. EXPERIMENTAL RESULTS

Single-crystalline platelets were selected from a high-purity polycrystalline ingot obtained in an induction furnace and annealed for two days at 1200 °C. X-ray diffraction and scanning electron microscope analysis revealed no traces of parasitic phases.[16] The samples were mounted in a tungsten-carbide anvil cell using steatite as pressure transmitting medium and a pyrophyllite gasket. Standard four-wires resistivity measurements were carried out in the basal plane of CePd$_2$Si$_2$ with a dc technique in a dilution refrigerator down to temperatures of 30 mK.

Resistivity curves versus temperature are shown in Fig. 1 for several pressures. At low pressure, the antiferromagnetic order is marked by a kink at T_N (at around 10 K) in the resitivity. This anomaly shifts to lower temperatures with increasing pressures. The extrapolation of the pressure variation of T_N to zero temperature defines the QCP at about 3.5 GPa. At low temperatures, a 10% drop of the resitivity is observed below 300 mK at 1.8 GPa (shown at 2.5 GPa in Fig. 1 for clarity). On increasing pressure this drop reaches 40% at 4.1 GPa and is followed by a saturation of the resitivity at the lowest temperatures. Following the results of the Cambridge group who found zero resitivity around these pressures, the drop is associated with the entrance in the

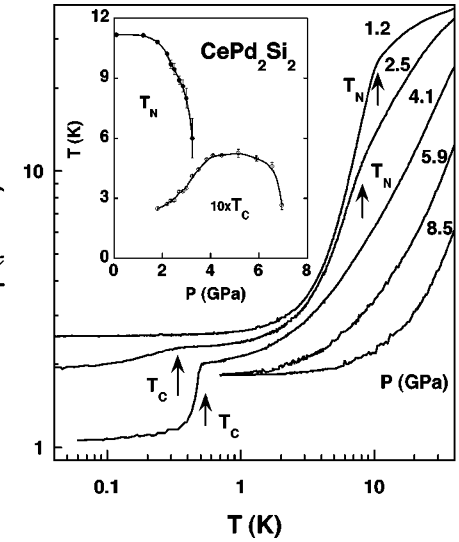

FIG. 1. Basal plane resistivity of CePd$_2$Si$_2$ versus temperature on a double logarithmic scale for several pressures. Arrows indicates the transition temperatures. The inset shows the $T-P$ phase diagram obtained.

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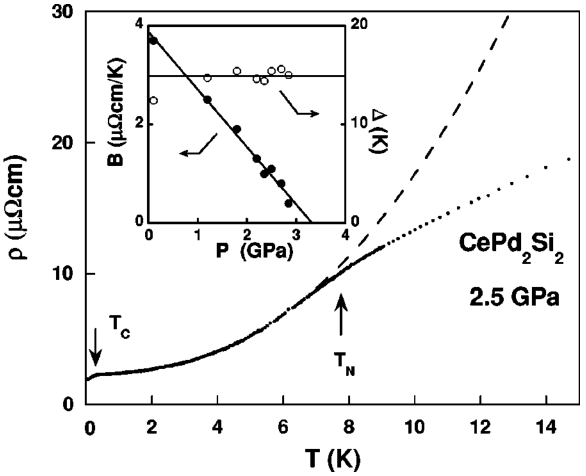

FIG. 2. Basal plane resistivity of CePd$_2$Si$_2$ at 2.5 GPa showing both a kink at $T_{\rm N}$ and a drop at $T_{\rm c}$. The dashed line is a fit to Eq. (1) given in the text. The inset shows the pressure variation of the spin-wave scattering weight B and the spin-wave energy gap Δ.

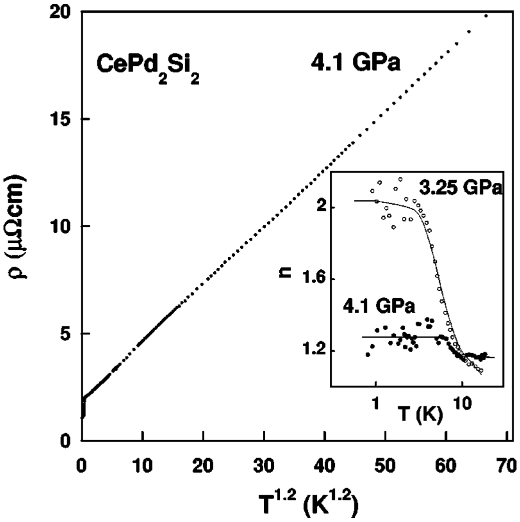

FIG. 3. Basal plane resistivity of CePd$_2$Si$_2$ at 4.1 GPa versus $T^{1.2}$. The inset shows the exponent n of the T^n resistivity law versus temperature at 3.25 and 4.1 GPa.

superconducting state. For pressures higher than 5.15 GPa, the drop in the resistivity gradually decreases again and the transition is no more observed at 7.5 GPa. At low pressure, signs of magnetism and superconductivity coexist as shown, for example, in Fig. 1 for a pressure of 2.5 GPa where both a $T_{\rm N}$ and a $T_{\rm c}$ value can be defined. The corresponding $T-P$ phase diagram is shown in the inset of Fig. 1. The pressure corresponding to the QCP (3.5 GPa) is different from the optimum pressure for superconductivity found at around 5 GPa.

In the antiferromagnetic phase, the resistivity between $T_{\rm c}$ and $T_{\rm N}$ is described with the following form previously used in the same context for the antiferromagnetic HF superconductor URu$_2$Si$_2$:[17]

$$\rho=\rho_0+AT^2+BT(1+2T/\Delta)\exp(-\Delta/T). \tag{1}$$

The first term, ρ_0, corresponds to the residual resistivity, the second term corresponds to the Fermi-liquid contribution of heavy electrons, and the last term to the contribution of antiferromagnetic gapped spin waves. A fit of Eq. (1) to the data is shown in Fig. 2 for a pressure of 2.5 GPa. The results obtained at low pressures show that the fit is not overparametrized. At 0.1 GPa, we found $A=0.098\ \mu\Omega\,\mathrm{cm\,K^{-2}}$ which is indeed consistent with the low-temperature specific-heat linear coefficient $\gamma\approx100$ mJ/mol K^2,[18,19] given a so-called Kadowaki-Woods ratio of $9.8\times10^{-6}\ \mu\Omega\,\mathrm{cm\,K^2(mol/mJ)^2}$ very close to the canonical value $10^{-5}\ \mu\Omega\,\mathrm{cm\,K^2(mol/mJ)^2}$. The estimate of the spin gap $\Delta=14$ K is close to the value of 9.6 K obtained by inelastic neutron scattering.[20] The pressure variation of the gap Δ and of the magnon scattering weight B are shown in the inset of Fig. 2. The gap is constant overall the pressure range and B decreases linearly and vanishes at the QCP (around 3.2–3.5 GPa). The fact that the gap is constant, is an indication that among the two components of the magnetic excitation spectrum,[20] namely a spin wave and a quasielastic part, only the latter is relevant and may play an important role in the driving mechanism of the QCP.

NFL behavior is observed in a narrow pressure range around the optimum pressure for superconductivity. This is shown in Fig. 3 for a pressure of 4.1 GPa. The resistivity follows a $T^{1.2}$ law up to 35 K. The same behavior is observed at 4.5 GPa. On the other hand, at the QCP (3.2–3.5 GPa), a Fermi-liquid-like T^2 law is observed for the resistivity below 3 K. Both behaviors are shown in the inset of Fig. 3 where the exponent n of the T^n law of the resistivity [obtained by the temperature logarithmic derivatives of $\rho(T)-\rho_0$] is plotted versus temperature. At 3.25 GPa, a large crossover regime is observed in the resitivity exponent between 10 and 3 K followed by a saturation towards $n=2$. On the other hand, n is almost constant over two decades in temperatures at 4.1 GPa. For higher pressures starting at 5.15 GPa, a T^2 law with a decreasing weight is again observed. Our results emphasize the fact that NFL behavior, observed in a narrow pressure window, seems to be better linked to the optimum superconductivity rather than to the QCP.

The initial slope of the superconducting upper critical field H_{c2} at $T_{\rm c}$ allows to give more insight in the electronic properties (effective mass) of HF compounds.[21] The field variation of the superconducting resistive transition at 3.5 GPa is shown in Fig. 4. On increasing the field, the resistivity drop shifts to lower temperatures and is no more seen for a field of 1.9 T. The corresponding $T-H$ phase diagram is shown in the inset of Fig. 4. The striking feature is the high initial slope of the critical field of about -13 T/K at this pressure. This high value is taken as an evidence for HF superconductivity since in the clean limit (reached here due to the low residual resitivity), the initial slope of the critical field is directly proportional to the square of the effective mass m^* of the quasiparticles forming the Cooper pairs. The pressure variation of the mass thus deduced and arbitrarily normalized to 1 at $P=2.2$ GPa is shown in the lower frame of Fig. 5. Another estimate of m^* is obtained from the square root of the A coefficient of the T^2 law of resistivity ($m^*\approx\sqrt{A}$). Its pressure variation is shown in the same figure. Between 2 and 6 GPa there is an agreement for the reduction (by a factor of about 2) of the effective mass deduced from this two quantities even if the ''path'' taken is

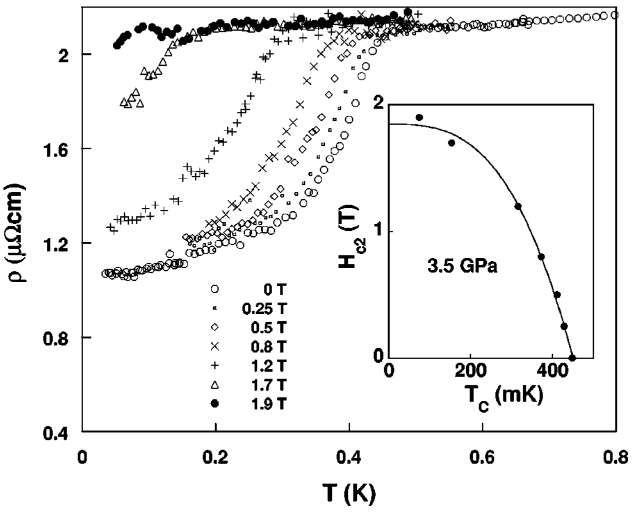

FIG. 4. Basal plane resistivity of $CePd_2Si_2$ at 3.5 GPa for different magnetic fields. The inset shows the $T-H$ phase diagram obtained. The line is a guide for the eye.

different. Magnetic (quasielastic) fluctuations may be part of the A coefficient in the magnetically ordered phase given thus an overestimated value of the effective mass. To this respect low-temperature specific-heat measurements under pressure will be very valuable to have a direct estimation of the effective mass. The pressure variation of the residual resitivity is shown by comparison in the upper frame of Fig. 5. This quantity, starting from 2.8 $\mu\Omega$ cm at $P=0$, decreases initially with pressure, equals 2.4 $\mu\Omega$ cm when superconductivity is observed and almost saturates at 1.8 $\mu\Omega$ cm after the pressure for optimal superconductivity, i.e., above 5 GPa. No scaling is found between ρ_0 and $\sqrt{A}$ which may indicate that the scattering given rise to this residual resitivity is nearly ''textbook'' defect scattering without any obvious contribution from Kondo physics (e.g., absence of ''Kondo-hole'' scattering mechanism).

III. DISCUSSION

Compared to a previous study of the pressure-induced superconductivity of $CePd_2Si_2$ observed in the range 2–3 GPa,[12] we found a larger pressure domain of existence of this phase between 2 and 7 GPa. Albeit surprising the observation of such a large domain is consistent with previous measurements performed on $CeCu_2Si_2$ in the same conditions[22] where superconductivity is found in the range 0–10 GPa. Such a similarity is more or less expected from the ''unified'' behavior of these 1-2-2 compounds as described in the introduction. Moreover, a better comparison can be made with the isoelectronic compound $CeNi_2Ge_2$. With a smaller unit cell, $CeNi_2Ge_2$ at zero pressure is believed to be the equivalent of $CePd_2Si_2$ at around 3 GPa. To this respect the observation of superconductivity in $CePd_2Si_2$ at 3 GPa and in $CeNi_2Ge_2$ at zero pressure is consistent with this picture. The other pocket of superconductivity observed in $CeNi_2Ge_2$ in the range 2–4 GPa (Ref. 11) could also be linked to the superconductivity we still observed at higher pressure (up to 7 GPa) in $CePd_2Si_2$. It is as if the large superconducting pressure range of $CePd_2Si_2$ we found in this work covers the two pockets observed in $CeNi_2Ge_2$ with a shift of pressure of the order of 3 GPa compensating the volume difference between the two compounds.

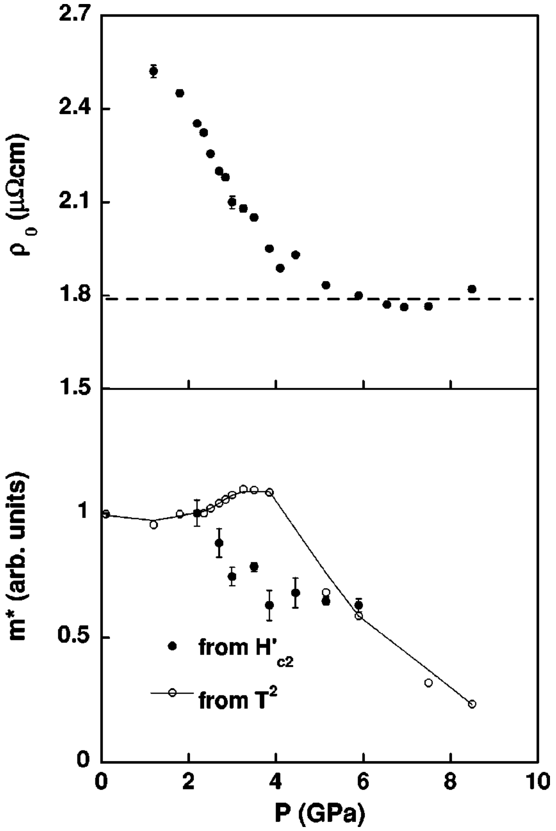

FIG. 5. Pressure variation of the residual resistivity (upper frame). Pressure variation of the effective mass (lower frame) deduced from the initial slope of the critical field and the A coefficient of the T^2 resistivity law, respectively. The line is a guide for the eyes.

A consequence of the large superconducting domain observed is the separation between the QCP and the pressure for optimal superconductivity to which NFL properties seems to be better linked. This bears striking similarities with the marginal behavior observed at the optimal doping in the high-temperature superconductors (HTSC's) where the resitivity is linear in T over decades in temperatures.[23] The proximity of both systems (HF and HTSC) to antiferromagnetism is the basis of theoretical works aiming to explain their resitivity.[24,25] In nearly antiferromagnetic compounds, part the scattering is due to hot spot, that is, portions of the Fermi surface spanned by **Q**, the antiferromagnetic wave vector. This is the main scattering process taken into account in the spin-fluctuation theory[3] where the resitivity follows a $T^{3/2}$ law for three-dimensional antiferromagnet (respectively a T law in two dimensions) at the QCP for $T\approx 0$. Beyond this behavior, most of the Fermi surface (i.e., portions not connected by **Q**) must give rise to a Fermi-liquid-like behavior which may short circuit the hot spot scattering. It was shown that disorder enhances hot spot scattering and for a three-dimensional antiferromagnet (HF case),[24] the competition between a T^2 resistivity and a $x+T^{3/2}$ one (where x is proportional to the impurity concentration) could give rise to crossover exponents close to 1 due to a breakdown of Mathiessen's rule. This theory[24] goes one step beyond the spin-fluctuation theory by taking into account disorder which changes the weight between the hot spot scattering and the conventional Fermi-liquid scattering.

Despite differences in the superconducting $T-P$ phase diagram obtained in this study and in that of Ref. 12, we measured the same power law in the resitivity around the QCP which indicates, in the framework presented above, a similar sample quality [with a residual resitivity ratio (RRR) of the order of 20]. It was found by the Cambridge group that samples with a RRR of the order of 60 gave an exponent close to 1[10]. Similar results were obtained for the isostructural compound $\mathrm{CeNi_2Ge_2}$.[8] The resistivity was fitted in a narrower temperature range (up to 3 K) at zero pressure with an exponent between 1.37 and 1.5 depending on the sample quality (the lower exponent, the better sample).

IV. CONCLUSION

Our resitivity measurements confirm the pressure-induced superconductivity of $\mathrm{CePd_2Si_2}$ observed by the Cambridge group. A larger domain (2–7 GPa) for superconductivity was found in the present study. This bears similarities with the isoelectronic compound $\mathrm{CeNi_2Ge_2}$, where two pockets of superconductivity were observed at around 0 and 3 GPa. A high initial slope of the superconducting upper critical field as well as NFL properties observed near the QCP suggest HF superconductivity and importance of soft spin fluctuations.

Note added in proof. We recently received a report from the Grenoble group of the observation of a complete superconducting transition at 2.5 GPa in a sample of the same batch as the one used in the present work.

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